Phase Transition in a Conserved-Mass Model of Aggregation and Dissociation

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We introduce a new model of aggregation of particles where in addition to diffusion and aggregation upon contact, a single unit of mass can dissociate from a conglomerate. This dissociation move conserves the total mass and leads to a striking behaviour in the steady state. As the parameters are varied, the system undergoes a dynamical phase transition in all dimensions. In one phase the mass distribution decays exponentially for large mass whereas in the other phase there is a power law distribution of masses which coexists with an infinite mass aggregate. The model is investigated analytically within mean field theory, and numerically in one dimension.

Conservation laws satisfied by the dynamics are known to modify drastically [1] the time-dependent behaviour of systems which are in thermal equilibrium. There is, however, a wide variety of inherently *nonequilibrium* systems in nature whose steady states are not described by the Gibbs distribution, but are determined by the dynamics itself. Examples include systems exhibiting self-organized criticality [2], several reaction-diffusion systems [3] and fluctuating interfaces [4]. Then the question naturally arises: What is the role played by conservation laws in selecting the steady state of such a nonequilibrium system? The question is particularly significant because of the propensity of many nonequilibrium systems to organize themselves into critical states, which are especially sensitive to such conditions. How do conservation laws modify the power laws characteristic of such critical states? Can they bring in completely new types of behaviour?

We address these questions for the steady states of an important class of nonequilibrium processes, namely those involving the twin phenomena of aggregation and diffusion. These processes are ubiquitous in nature, and arise in a variety of physical settings, for instance, in the formation of colloidal suspensions [5] and polymer gels [6] on the one hand, and aerosols and clouds [7] on the other. A recent interesting and important result in this area due to Takayasu and collaborators [8], which has widespread applications, is that constant injection of single particles into such a system leads to a power law distribution of particle masses in the steady state. This injection of particles from outside of course violates mass conservation. In this Letter, we show that a conserved-mass system in which injection is replaced by *dissociation*, exhibits strikingly different and even more interesting behaviour: the new dissociation moves that conserve the total mass induce a novel dynamical phase transition. As the parameters of the system are changed, there is a transition from one steady state where the mass distribution decays exponentially to another where it decays as a power law, and in addition develops an infinitely large aggregate.

The conserved-mass aggregation model (CMAM) discussed here has connections to models of gelation [6] and to the Takayasu model (TM) of particle injection alluded to above [8]. However it turns out to have a steady state structure which is richer than either. In experiments [9] as well as theoretical models [6] of irreversible gel formation, the steady-state mass distribution $P(m)$ is trivial, as there is only a single infinite aggregate, though the kinetics of the approach to this state is quite interesting [6]. In the TM on the other hand the injection move completely destroys the infinite aggregate and the steady state mass distribution $P(m)$ decays as a power law $m^{-\tau}$ for large mass, where the exponent τ depends on the spatial dimension.

Our conserved-mass model differs from conventional models of aggregation in that single particles are allowed to chip off from more massive conglomerates. This move corresponds to the physical process of single functional units breaking off from larger clusters in the polymerization problem. It leads to a replenishment of the lower end of the mass spectrum, and competes with the tendency of the coalescence process to produce more massive aggregates. The result of this competition is that two types of steady states are possible, and there is a dynamical phase transition between the two. In one state, $P(m)$ decays exponentially, while the other is more interesting: $P(m)$ decays as a power law for large m but in addition develops a delta function peak at $m = \infty$. Physically this means that an infinite aggregate forms that subsumes a finite fraction of the total mass, and coexists with smaller finite clusters whose mass distribution has a power law tail. In the language of sol-gel transition, the infinite aggregate is like the gel while the smaller clusters form the sol. However, as opposed to the models of irreversible gelation where the sol disappears in the steady state, in our model the sol coexists with the gel even in the steady state. Interestingly, the mechanism of the formation of the infinite aggregate in the steady state resembles Bose-Einstein condensation (BEC), though the condensate (the infinite aggregate here) forms in real space rather than momentum space as in conventional BEC.

Our model can be considered as the conserved counterpart of the non-conserved TM [8]. The injection move in TM that violates the mass conservation is replaced in our model by the dissociation move that conserves the mass. Besides being a simple model having a self-organized critical state, TM has found widespread applications including modeling of river networks [10] and stress distribution

in granular media [11]. It is therefore not unreasonable to expect that the conserved mass model discussed here will also find applications in a wide variety of physical processes. As an example, it can be considered as a simple model of river networks in a basin where there is negligible rainfall (injection) but small rivulets can break off a stream (dissociation).

The CMAM is defined as follows. For simplicity we define the model on a one dimensional lattice with periodic boundary conditions although generalizations to higher dimensions are quite straightforward. Beginning with a state in which the masses are placed randomly, a site is chosen at random. If it contains one or more than one particle, then one of the following events can occur:

1. Diffusion and Aggregation: With probability p_1, the mass m_i at site i moves either to site $i-1$ or to site $i+1$. If it moves to a site which already has some particles, then the total mass just adds up.
2. Chipping (single-particle dissociation): With probability p_2, a bit of the mass at the site "chips" off, *i.e.* a single particle leaves site i and moves with equal probability to one of the neighbouring sites $i-1$ and $i+1$.
3. With a probability $1-p_1-p_2$, the site is left undisturbed.

If the site chosen is empty, then nothing happens. The same rules hold even if we choose a site with only a single particle, which means that the probability for a single particle to move left or right in this model is p_1+p_2. Note the difference with TM: in TM, the move 2 is replaced by addition of unit mass to every site with probability 1.

We first analyze the model within the mean field approximation, ignoring correlations in the occupancy of adjacent sites. Then we can directly write down equations for $P(m,t)$, the probability that any site has a mass m at time t.

$$\frac{dP(m,t)}{dt} = -(p_1+p_2)[1+q(t)]P(m,t) + p_2P(m+1,t) + p_2q(t)P(m-1,t) + p_1P*P; \quad m \geq 1 \qquad (1)$$

$$\frac{dP(0,t)}{dt} = -(p_1+p_2)q(t)P(0,t) + p_2P(1,t) + p_1q(t). \qquad (2)$$

Here $q(t) \equiv 1-P(0,t)$ is the probability that a site is occupied by a mass and $P*P = \sum_{m'=1}^{m} P(m',t)P(m-m',t)$ is a convolution term that describes the coalescence of two masses.

The above equations enumerate all possible ways in which the mass at a site might change. The first term in Eq. (1) is the "loss" term that accounts for the probability that a mass m might move as a whole or chip off to either of the neighbouring sites, or a mass from the neighbouring site might move or chip off to the site in consideration. The probability of occupation of the neighbouring site, $q(t) = \sum_{m=1} P(m,t)$, multiplies $P(m,t)$ within the mean-field approximation where one neglects the spatial correlations in the occupation probabilities of neighbouring sites. The remaining three terms in Eq. (1) are the "gain" terms enumerating the number of ways that a site with mass $m' \neq m$ can gain the deficit mass $m-m'$. The second equation Eq. (2) is a similar enumeration of the possibilities for loss and gain of empty sites. Evidently, the mean field equations conserve the total mass.

To solve the equations, we compute the generating function, $Q(z,t) = \sum_{m=1}^{\infty} P(m,t)z^m$ from Eq. (1) and set $\partial Q/\partial t = 0$ in the steady state. We also need to use Eq. (2) to write $P(1,t)$ in terms of $q(t)$. This gives us a quadratic equation for Q in the steady state. Choosing the root that corresponds to $Q(z=0)=0$, we find

$$Q(z) = \frac{w+2q+wq}{2} - \frac{w}{2z} - \frac{wqz}{2} + wq\frac{(1-z)}{2z}\sqrt{(z-z_1)(z-z_2)}. \qquad (3)$$

where $w = p_2/p_1$ and $z_{1,2} = (w+2\mp 2\sqrt{w+1})/wq$. The value of the occupation probability q is fixed by mass conservation which implies that $\sum mP(m) = M/L \equiv \rho$. Putting $\partial_z Q(z=1) = \rho$, the resulting relation between ρ and q is

$$2\rho = w(1-q) - wq\sqrt{(z_1-1)(z_2-1)}\ . \qquad (4)$$

The steady state probability distribution $P(m)$ is the coefficient of z^m in $Q(z)$ and can be obtained from $Q(z)$ in Eq. (3) by evaluating the integral

$$P(m) = \frac{1}{2\pi i}\int_{C_o} \frac{Q(z)}{z^{m+1}}dz \qquad (5)$$

over the contour C_o encircling the origin. The singularities of the integrand govern the asymptotic behaviour of $P(m)$ for large m. Clearly the integrand has branch cuts at $z = z_{1,2}$. For fixed w, if one increases the density ρ, the occupation probability q also increases as evident from Eq. (4). As a result, both the roots $z_{1,2}$ start decreasing. As long as the lower root z_1 is greater than 1, Eq. (4) is well defined and the analysis of the contour integration around the branch cut $z = z_1$, yields for large m,

$$P(m) \sim e^{-m/m^*}/m^{3/2}\ , \qquad (6)$$

where the characteristic mass, $m^* = 1/\log(z_1)$ and diverges as $\sim (q_c-q)^{-1}$ as q approaches $q_c = (w+2-2\sqrt{w+1})/w$. q_c is the critical value of q at which $z_1 = 1$. This exponentially decaying mass distribution is the signature of "disordered" phase which occurs for $q < q_c$ or equivalently from Eq. (4) for $\rho < \rho_c = \sqrt{w+1}-1$.

When $\rho = \rho_c$, we have $z_1 = 1$, and analysis of the contour around $z = z_1 = 1$ yields a power law decay of $P(m)$,

$$P(m) \sim m^{-5/2}. \tag{7}$$

As ρ is increased further beyond ρ_c, q can not increase any more because if it does so, the root z_1 would be less than 1 (while the other root z_2 is still bigger than 1) and Eq. (4) would be undefined. The only possibility is that q sticks to its critical value q_c or equivalently the lower root z_1 sticks to 1. Physically this implies that adding more particles does not change the occupation probability of sites. This can happen only if all the additional particles (as ρ is increased) aggregate on a vanishing fraction of sites, thus not contributing to the occupation of the others. Hence in this "infinite-aggregate" phase $P(m)$ has an infinite-mass aggregate, in addition to the power law decay $m^{-5/2}$. Concomitantly Eq. (4) ceases to hold, and the relation now becomes

$$\rho = \frac{w}{2}(1 - q_c) + \rho_\infty \tag{8}$$

where ρ_∞ is the fraction of the mass in the infinite aggregate. The mechanism of the formation of the aggregate is reminiscent of Bose Einstein condensation. In that case, for temperatures in which a macroscopic condensate exists, particles added to the system do not contribute to the occupation of the excited states; they only add to the condensate, as they do to the infinite aggregate here.

Thus the mean field phase diagram (Fig. 1) of the system consists of two phases, "disordered" and "infinite-aggregate", which are separated by the phase boundary, $\rho_c = \sqrt{w+1} - 1$. While this phase diagram remains qualitatively the same even in 1-d, the exponents characterizing the power laws are different from their mean field values (see Fig. 1).

We have studied this model using Monte Carlo simulations on a one-dimensional lattice. Although we present results here for a relatively small size lattice, $L = 1024$, we have checked our results for larger sizes as well. We confirmed that all the qualitative predictions of the mean-field theory remain true, by calculating $P(m)$ numerically in the steady state. Figure 2 displays two numerically obtained plots of $P(m)$. The existence of both the disordered (denoted by $+$) and the infinite-aggregate phase (denoted by $\times$) is confirmed. In particular, the second curve shows clear evidence of a power-law behaviour of the distribution, which is cut off by finite-size effects, and for an 'infinite' aggregate beyond. We confirmed that the mass M_{agg} in this aggregate grows linearly with the size, and that the spread δM_{agg} grows sublinearly, implying that the ratio $\delta M_{agg}/M_{agg}$ approaches zero in the thermodynamic limit. The exponent τ which characterizes the finite-mass fragment power law decay is numerically found to be $2.33 \pm .02$. The difference of this value from the mean-field value 2.5 is presumably due to the neglect of spatial correlations within mean-field theory.

We note that in TM, the exponent τ_{TM} that characterizes the power law decay of mass distribution in the steady state has an exact value 4/3 in 1-d and 3/2 within mean field theory [8]. In the CMAM, we find that in the aggregate phase $\tau_{CMAM} \sim 2.33$ in 1-d and 5/2 within the mean field theory. It is therefore tempting to conjecture that $\tau_{CMAM} = \tau_{TM} + 1$ although we have no proof of this.

Mass conservation evidently affects the steady state in this class of nonequilibrium models, but what are the other factors which determine the universality classes? We addressed this question by studying the effect of directionality on the motion of the masses, leading to a finite mass current. For the mass-nonconserved TM, it is known that making such a change has no effect on the scaling properties [8]. Our numerical study of the directed mass-conserved case shows [13] that directionality in fact changes the universality class. As in the undirected case, the model continues to show a phase transition between two phases, without and with an infinite aggregate, respectively. However, the exponent τ characterizing the power law decay in the infinite-aggregate phase, is different in this case (Fig. 3). In this model, $\tau \simeq 2.05$ within numerical error [13]. Clearly, $P(m)$ should decay faster than m^{-2} for large m to keep the total mass $\sum mP(m)$ finite. Perhaps $P(m)$ decays as m^{-2} with additional logarithmic factors.

Interestingly, the CMAM can be mapped onto a driven diffusive lattice gas model, and thence to a model of interface dynamics, so that our results for aggregation phenomena have wider significance. A configuration of the CMAM is mapped onto a particle-hole configuration as follows: Every site with m particles in the aggregation model is mapped to a cluster of m successively occupied sites, with a vacancy at the rightmost edge in the particle model. An empty site maps to an empty site. Thus a lattice of L sites in the aggregation-dissociation model maps to one of $L + N_p$ sites in the hard-core particle model, where N_p is the total number of particles. The dynamics defined in the earlier section now translates to the following rules for particle motion. If a randomly selected site is occupied, then with probability p_1, the entire particle cluster to which the occupied site belongs is moved one site to the left or right. With probability p_2, the rightmost or leftmost particle from the cluster dissociates from the cluster and moves one site to the right or left. With probability $1 - p_1 - p_2$, the system is left undisturbed. The mapping to an interface model follows a standard route [12]: a particle is mapped to a unit segment with a positive slope, while a hole maps to a segment of unit length with a negative slope. The equivalent interface then allows both for single corner flips at hills and valleys, and also for slice-wise moves of connected segments of up and down slopes. Under this dynam-

ics, interface profiles can develop very different spatial structures as compared to those described in customary growth processes [4]. In the exponential phase, particle cluster lengths decay exponentially as is usual, but in the infinite-aggregate phase, a single macroscopically long stretch with positive slope develops. This unusual behaviour differs from descriptions of mound formation [14], in that the infinite stretch is accompanied by the formation of fairly long finite stretches with a power-law distribution of stretch lengths. This defines a new universality class in interface dynamics.

In conclusion, we point out that several questions still remain open. Amongst these is the understanding of spatial and temporal correlations, and the role of directionality and dimensionality in influencing the scaling behaviour, in particular, the determination of the upper critical dimension.

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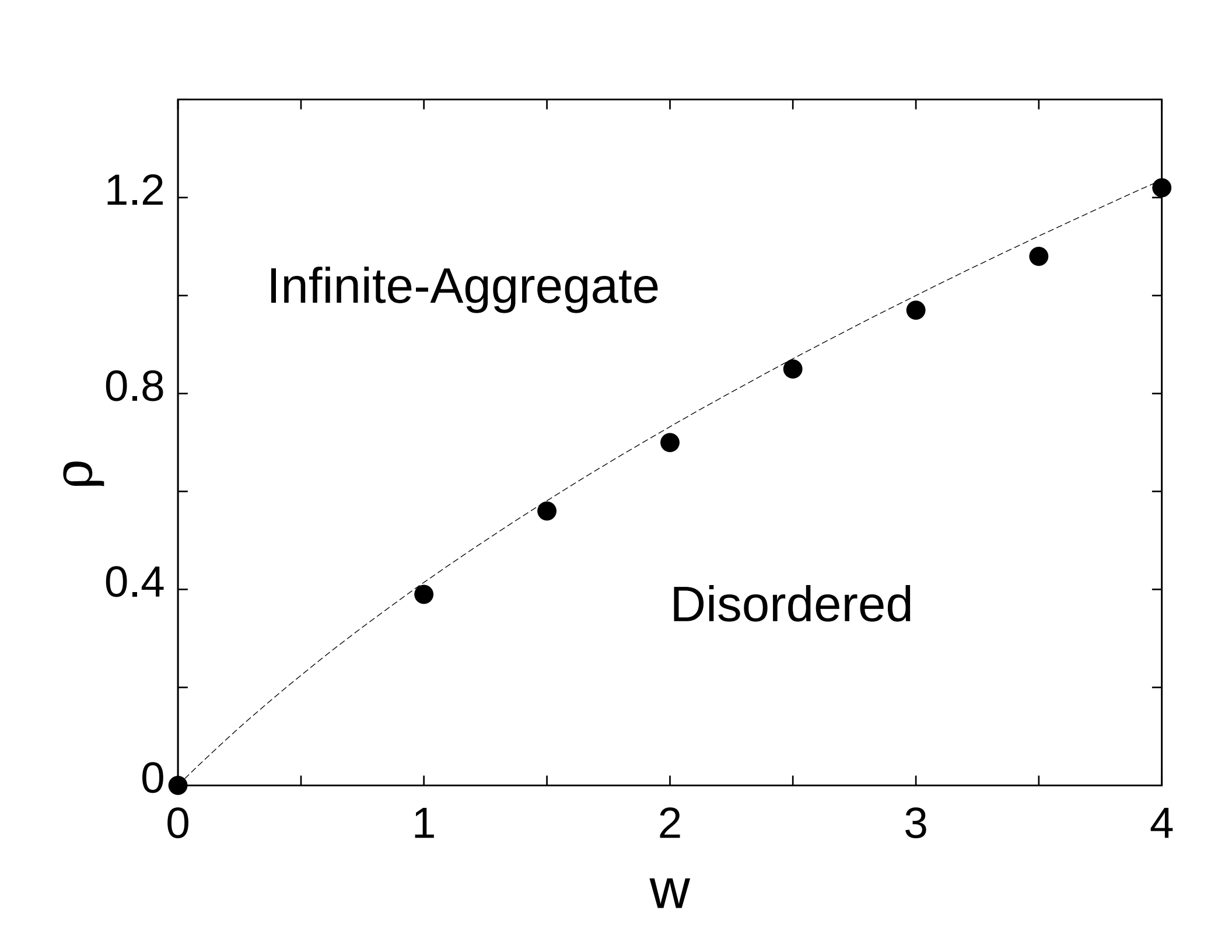
Figure Captions

Fig. 1. The phase diagram in the ρ-w plane. The dashed line shows the mean field phase boundary. The points denoted by ($\bullet$) are obtained numerically in one dimension.

Fig. 2. The mass distribution $P(m)$ vs. m for the undirected model in 1-d on a log-log plot for $w = 1.0$ and $\rho = 0.2$ (shown by $\times$ symbols) and for $w = 1.0$ and $\rho = 3.0$ (shown by + symbols).

Fig. 3. The mass distribution $P(m)$ vs. m (only the power law part) on a log-log plot for both undirected (+) and directed ($\times$) case in the "Infinite-Aggregate" phase at density $\rho = 12.0$ and $w = 1.0$. The straight line with slope -2 is drawn as a guidance to the eyes.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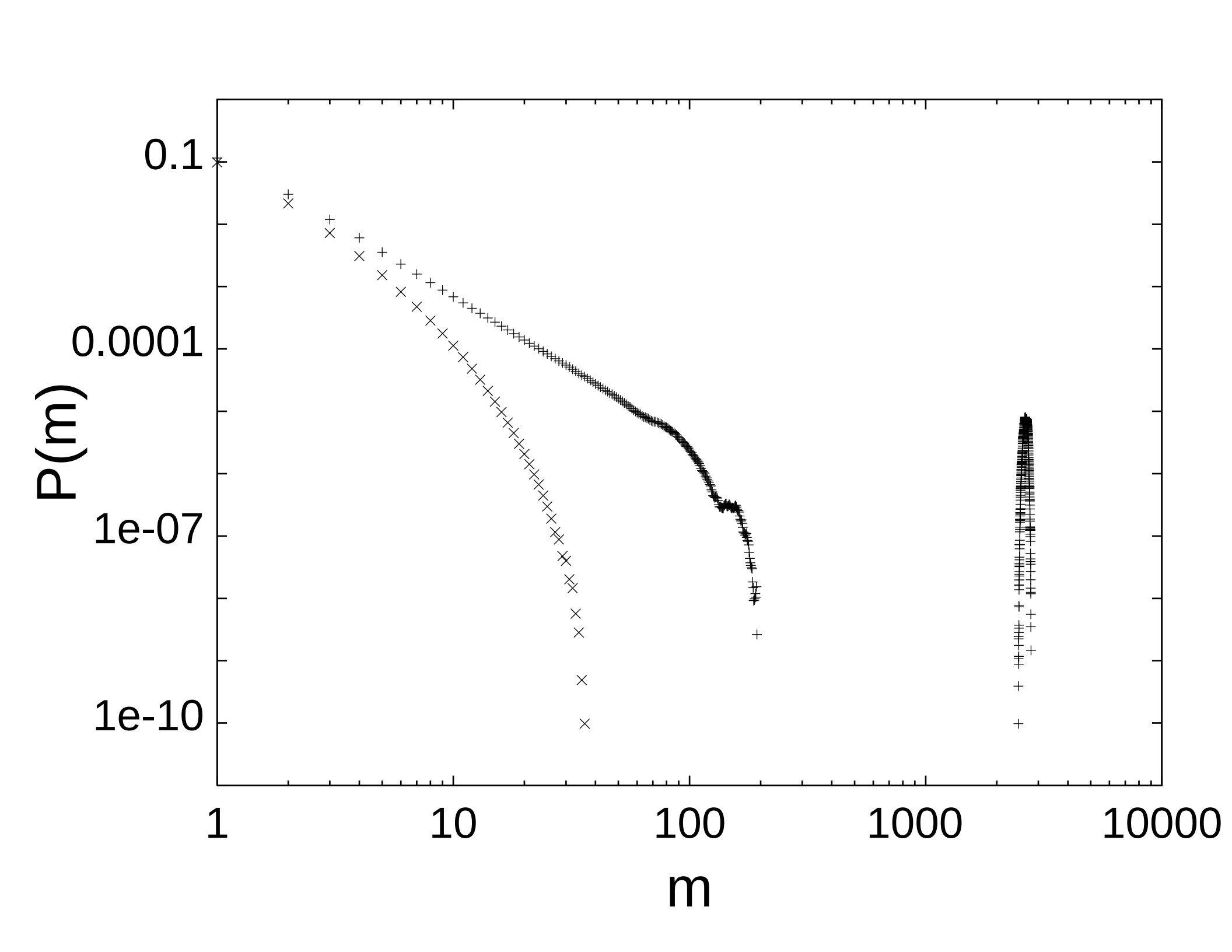
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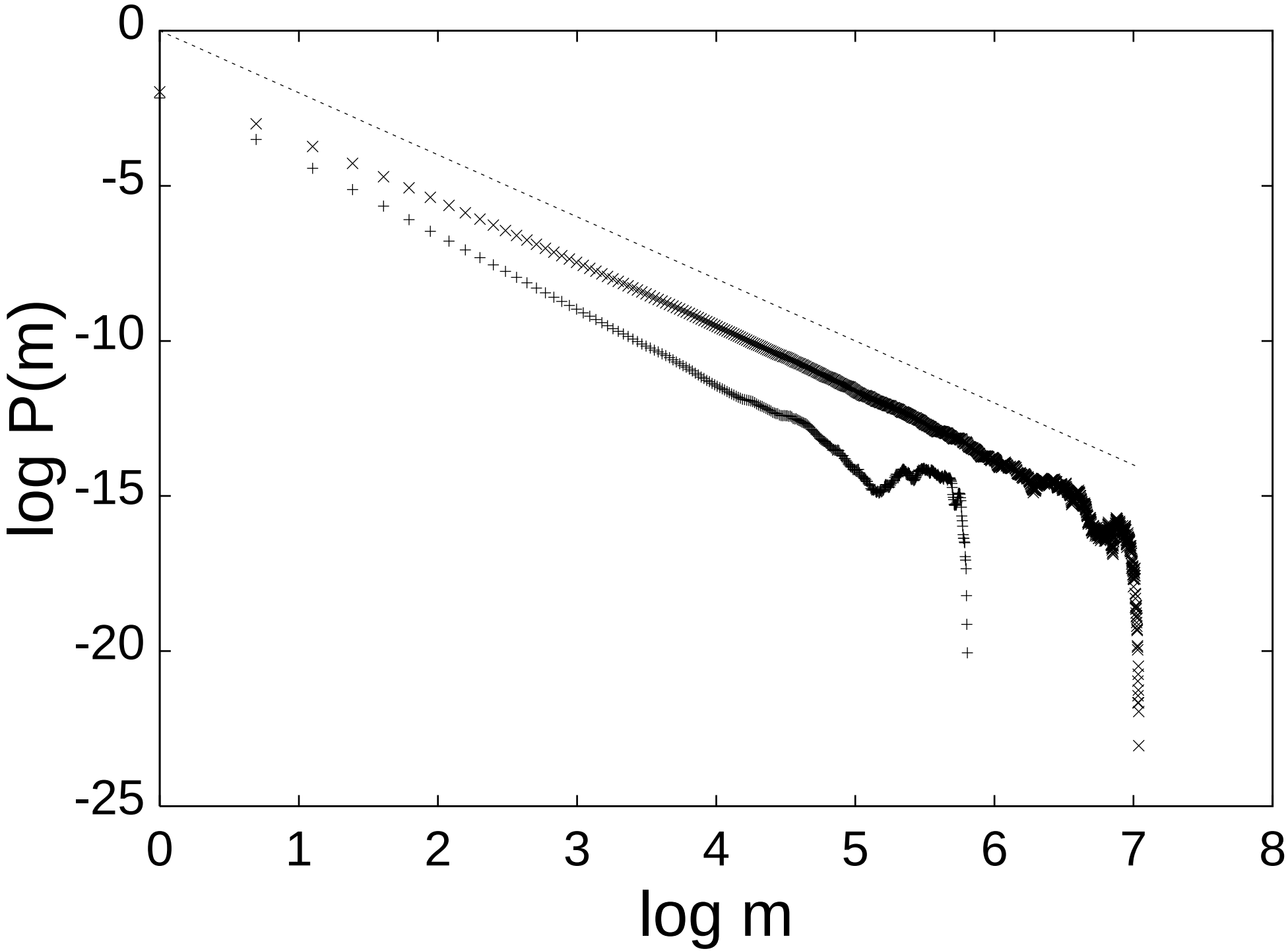
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log P(m)
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